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**Supporting information for article:**

**Quantification of amorphous siliceous fly ash in hydrating blended cement pastes by X-ray powder diffraction**

**Xuerun Li, Ruben Snellings and Karen L. Scrivener**

## ***Supplementary Material for***

### **Quantification of amorphous siliceous fly ash in hydrating blended cement pastes by X-ray powder diffraction**

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#### ***1. The phase assemblage.***

Table S1: Phase assemblage of the samples in the unit of g / (100 g of paste). Results of the **dried powder**, Rietveld analysis using **13.5 – 70 degrees** of XRD patterns.

Table S2: Phase assemblage of the samples in the unit of g / (100 g of paste). Results of the **fresh discs**, Rietveld analysis using **7 – 70 degrees** of XRD patterns.

Notes for the abbreviations used in the tables:

Hemicarb. = hemicarboaluminate, Monocarb. = monocarboaluminate, AFm = monosulfate, Am.FA = amorphous fly ash, CH = portlandite, AFt = ettringite.

#### ***2. Calculation process.***

Table S3: The calculation process of the heat release from FA based on the DoH of clinker and the calorimetry results was also given.

Figure S1: The fitting of the FA\_30 at 90 days, Rietveld analysis was carried out starting from 7 degrees.

Figure S2: The Rietveld plot of the cement with a wide 2 theta angle

Figure S3: 7-year white cement, C-S-H profile modelled using 4 peaks.

Figure S4: 90d white cement, C-S-H profile from 7-year old sample.

Figure S5: 90d white cement, crystal size of peak at 0.3037 nm was refined.

#### ***3. Bound water.***

Table S4: The bound water detected by TGA

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Table S1 Phase assemblage of the samples in the unit of g / (100 g of paste). Results of the dried powder, Rietveld analysis using 13.5 – 70 degrees of XRD patterns.

Sample	Time (d)	C <sub>3</sub> S	C <sub>2</sub> S	C <sub>3</sub> A	C <sub>4</sub> AF	Lime	Periclase	Anhydrite	Magnetite	Quartz	Aft	CH	Hemicarb.	Monocarb.	AFm	Mullite	Arcanite	Gypsum	Am. FA
PC	0	47.1	3.6	5.1	10.0	0.2	0.4	2.9	0.0	0.2	0.0	0.0	0.0	0.0	0.0	0.0	1.9	0.0	0.0
PC	1	17.7	1.3	3.1	7.5	0.0	0.2	0.0	0.0	0.1	2.8	8.5	0.2	0.4	0.8	0.0	0.0	0.0	0.0
PC	3	11.4	1.5	1.5	6.0	0.0	0.2	0.0	0.0	0.1	2.2	10.8	0.3	0.7	2.2	0.0	0.0	0.0	0.0
PC	7	9.2	1.3	1.2	5.5	0.0	0.2	0.0	0.0	0.1	1.8	11.2	0.8	0.7	2.0	0.0	0.0	0.0	0.0
PC	28	7.0	0.9	0.9	4.7	0.0	0.2	0.0	0.0	0.2	2.4	12.6	0.5	2.0	1.9	0.0	0.0	0.0	0.0
PC	90	5.3	0.7	0.9	4.5	0.0	0.0	0.0	0.0	0.2	2.1	13.8	0.7	2.1	2.0	0.0	0.0	0.0	0.0
PC	790	5.1	1.1	0.6	3.9	0.0	0.0	0.0	0.0	0.2	1.3	13.4	0.3	0.8	2.4	0.0	0.0	0.0	0.0
FA_10	0	42.1	3.2	4.6	8.9	0.2	0.3	2.6	0.0	1.1	0.0	0.0	0.0	0.0	0.0	1.3	1.7	0.5	4.8
FA_10	1	15.3	1.5	2.3	6.7	0.0	0.2	0.0	0.0	0.8	3.3	7.5	0.0	0.4	1.2	1.7	0.0	0.0	4.0
FA_10	3	8.6	1.2	1.1	5.3	0.0	0.1	0.0	0.0	1.1	3.2	10.7	0.0	0.7	2.3	1.6	0.0	0.0	2.9
FA_10	7	6.1	0.9	0.8	4.5	0.0	0.1	0.0	0.0	1.1	2.7	12.0	0.0	1.7	2.6	1.3	0.0	0.0	3.1
FA_10	28	4.5	1.0	0.7	4.0	0.0	0.2	0.0	0.0	1.0	2.9	11.8	0.0	1.4	1.9	1.4	0.0	0.0	2.1
FA_10	90	3.8	0.6	0.6	4.0	0.0	0.1	0.0	0.0	1.1	3.4	11.1	0.2	0.7	2.8	1.3	0.0	0.0	2.1
FA_10	790	3.1	1.3	0.4	3.3	0.0	0.1	0.0	0.0	1.0	3.0	11.4	0.0	2.3	1.9	1.1	0.0	0.0	1.9
FA_30	0	32.3	2.5	3.5	6.8	0.2	0.3	2.0	0.1	2.8	0.0	0.0	0.0	0.0	0.0	4.0	1.3	1.4	14.2
FA_30	1	9.8	0.9	1.8	5.0	0.0	0.1	0.0	0.0	2.7	3.2	6.0	0.0	0.5	0.6	4.5	0.0	0.0	14.1
FA_30	3	4.8	1.4	0.9	4.0	0.0	0.2	0.0	0.0	2.9	4.2	8.2	0.1	0.5	1.2	4.3	0.0	0.0	13.7
FA_30	7	3.0	0.9	0.6	3.3	0.0	0.1	0.0	0.0	2.8	3.4	8.9	0.0	1.8	1.6	4.3	0.0	0.0	13.2
FA_30	28	1.7	0.7	0.3	2.9	0.0	0.1	0.0	0.0	2.9	4.7	8.7	0.4	1.7	1.5	4.2	0.0	0.0	11.4
FA_30	90	1.6	0.4	0.2	3.0	0.0	0.2	0.0	0.0	3.0	5.9	7.8	0.0	0.7	2.2	4.2	0.0	0.0	11.0
FA_30	790	1.2	0.5	0.2	2.4	0.0	0.2	0.0	0.0	2.7	4.7	5.9	0.0	1.1	1.8	3.8	0.0	0.0	8.9
FA_50	0	22.8	1.7	2.5	4.8	0.1	0.2	1.4	0.2	4.4	0.0	0.0	0.0	0.0	0.0	6.5	0.9	2.3	23.4
FA_50	1	5.9	0.6	1.5	3.4	0.0	0.1	0.0	0.0	4.5	3.9	4.3	0.1	0.6	0.0	6.8	0.0	0.0	23.4
FA_50	3	2.3	0.9	0.7	2.9	0.0	0.2	0.0	0.0	4.5	6.0	5.3	0.2	1.0	0.1	6.8	0.0	0.0	22.8
FA_50	7	1.4	0.6	0.3	2.5	0.0	0.1	0.0	0.0	4.4	4.7	5.1	0.0	1.1	0.3	6.6	0.0	0.0	22.1
FA_50	28	0.7	0.5	0.1	2.1	0.0	0.1	0.0	0.0	4.4	5.7	4.7	0.0	1.2	0.8	6.7	0.0	0.0	19.7
FA_50	90	0.6	0.4	0.0	1.8	0.0	0.2	0.0	0.0	4.4	8.1	3.6	0.0	1.4	0.8	6.3	0.0	0.0	18.8
FA_50	790	0.3	0.3	0.0	1.6	0.0	0.2	0.0	0.0	4.6	6.6	2.9	0.0	1.0	0.6	6.5	0.0	0.0	17.3

Table S2 Phase assemblage of the samples in the unit of g / (100 g of paste). Results of **the fresh discs**, Rietveld analysis using **7–70 degrees** of XRD patterns.

Sample	Time (d)	C <sub>3</sub> S	C <sub>2</sub> S	C <sub>3</sub> Acubic	C <sub>3</sub> Aorth	C4AF	Lime	Periclase	Anhydrite	Magnetite	Quartz	Aft	CH	Hemicarb.	Monocarb.	AFm	Mullite	Arcanite	Gypsum
PC	0	47.1	3.6	1.2	4.0	10.0	0.2	0.4	2.9	0.0	0.2	0.0	0.0	0.0	0.0	0.0	0.0	1.9	0.0
PC	1	18.2	1.5	0.4	2.6	7.1	0.0	0.3	0.0	0.0	0.1	7.5	9.6	0.6	1.1	0.3	0.0	0.0	0.0
PC	3	13.1	1.3	0.2	1.6	6.5	0.0	0.2	0.0	0.0	0.1	8.1	11.5	0.6	1.8	0.6	0.0	0.0	0.0
PC	7	11.2	1.9	0.2	1.1	6.0	0.0	0.2	0.0	0.0	0.1	7.6	12.8	1.2	1.2	1.0	0.0	0.0	0.0
PC	28	8.4	1.6	0.3	0.6	5.2	0.0	0.2	0.0	0.0	0.1	7.8	14.0	0.5	1.2	2.0	0.0	0.0	0.0
PC	90	7.5	1.4	0.3	0.7	5.4	0.0	0.2	0.0	0.0	0.1	6.0	14.1	0.6	0.9	2.0	0.0	0.0	0.0
PC	790	5.0	1.2	0.0	0.7	3.3	0.0	0.1	0.0	0.0	0.2	6.2	13.9	0.0	3.3	4.1	0.0	0.0	0.0
FA_10	0	42.1	3.2	1.0	3.5	8.9	0.2	0.3	2.6	0.0	1.1	0.0	0.0	0.0	0.0	0.0	1.3	1.7	0.5
FA_10	1	15.6	0.8	0.5	2.2	6.3	0.0	0.3	0.0	0.0	1.5	7.9	8.3	0.7	0.8	0.3	1.8	0.0	0.0
FA_10	3	9.7	1.3	0.4	1.0	6.0	0.0	0.2	0.0	0.0	1.1	8.1	11.0	0.7	0.8	0.9	1.7	0.0	0.0
FA_10	7	8.6	0.9	0.3	0.6	5.1	0.0	0.1	0.0	0.0	1.0	5.8	11.4	0.8	0.2	0.0	1.8	0.0	0.0
FA_10	28	6.4	1.0	0.3	0.4	4.4	0.0	0.0	0.0	0.0	1.1	6.1	11.6	0.0	0.6	0.6	1.8	0.0	0.0
FA_10	90	3.9	1.3	0.1	0.4	3.6	0.0	0.3	0.0	0.0	1.2	9.3	12.2	0.0	1.1	3.2	1.5	0.0	0.0
FA_10	790	3.4	0.5	0.0	0.5	2.8	0.0	0.1	0.0	0.0	1.2	7.4	10.7	1.4	2.5	3.4	1.9	0.0	0.0
FA_30	0	32.3	2.5	0.8	2.7	6.8	0.2	0.3	2.0	0.1	2.8	0.0	0.0	0.0	0.0	0.0	4.0	1.3	1.4
FA_30	1	9.8	0.7	0.5	1.4	5.0	0.0	0.3	0.0	0.0	3.4	7.7	6.3	0.1	0.2	0.2	5.1	0.0	0.0
FA_30	3	5.3	0.9	0.2	0.7	3.9	0.0	0.1	0.0	0.0	3.3	9.4	9.1	0.4	0.9	0.6	4.7	0.0	0.0
FA_30	7	3.7	0.8	0.2	0.4	3.5	0.0	0.1	0.0	0.0	3.4	7.3	9.1	0.8	0.8	0.5	5.0	0.0	0.0
FA_30	28	2.3	0.5	0.1	0.2	2.7	0.0	0.1	0.0	0.0	3.2	7.2	10.0	0.2	0.8	0.8	4.8	0.0	0.0
FA_30	90	1.4	1.0	0.1	0.3	2.9	0.0	0.1	0.0	0.0	3.4	10.2	8.0	0.2	1.0	2.5	4.4	0.0	0.0
FA_30	790	1.1	0.9	0.0	0.1	2.4	0.0	0.3	0.0	0.0	3.1	8.7	7.9	0.0	0.9	2.2	4.3	0.0	0.0
FA_50	0	22.8	1.7	0.6	1.9	4.8	0.1	0.2	1.4	0.2	4.4	0.0	0.0	0.0	0.0	0.0	6.5	0.9	2.3
FA_50	1	6.5	0.6	0.3	0.9	3.4	0.0	0.2	0.0	0.0	4.7	5.1	4.6	0.2	0.7	0.0	7.8	0.0	0.0
FA_50	3	3.3	1.1	0.2	0.5	2.9	0.0	0.2	0.0	0.0	4.6	8.7	5.9	0.1	1.0	0.0	7.1	0.0	0.0
FA_50	7	1.6	0.7	0.0	0.4	2.4	0.0	0.3	0.0	0.0	5.1	9.7	5.6	0.3	1.5	0.2	7.3	0.0	0.0
FA_50	28	0.6	0.6	0.0	0.2	2.0	0.0	0.3	0.0	0.0	5.3	11.0	4.9	0.0	0.6	0.8	7.3	0.0	0.0
FA_50	90	0.6	0.4	0.0	0.0	1.9	0.0	0.3	0.0	0.0	5.0	11.3	4.3	0.0	0.6	0.8	7.2	0.0	0.0
FA_50	790	0.3	0.4	0.0	0.0	1.5	0.0	0.3	0.0	0.0	5.2	9.7	2.9	0.0	0.6	0.8	7.1	0.0	0.0

Table S3 Fly ash degree of reaction calculation from combined XRD and calorimetry data

Samples	Ages (d)	DoH (%)		Heat (J/g paste)		Heat (J/g SCM added)
		<i>of clinker (XRD)</i>	<i>from clinker</i>	<i>Detected by Calo.</i>	<i>from FA</i>	
PC	1	54.9	138			
	3	69	206			
	7	73.8	225			
	28	79.5	250			
Equation obtained from PC: Heat = 4.5623 × DoH – 111.42, R <sup>2</sup> = 0.999						
FA_10	1	56	128.78	122	-6.78	-95.63
	3	72.5	196.07	193	-3.07	-43.34
	7	79	222.58	214	-8.58	-121.00
	28	82.6	237.26	238	0.74	10.37
FA_30	1	61.1	114.79	102	-12.79	-60.92
	3	75.5	159.86	168	8.14	38.76
	7	82.7	182.40	194	11.60	55.26
	28	87.7	198.04	220	21.96	104.55
FA_50	1	64.1	87.54	70	-17.54	-50.79
	3	78.9	120.20	123	2.80	8.12
	7	84.9	133.43	152	18.57	53.75
	28	89	142.48	181	38.52	111.51

Table S4 The bound water detected by TGA

Time days	Bound water until 550 °C (wt. % in dried basis)			
	PC	FA_10	FA_30	FA_50
1	12.49	11.74	9.83	7.12
3	14.46	14.14	12.71	10.69
7	15.46	15.03	14.86	12.32
28	16.88	16.53	16.42	14.23
90	18.13	17.91	16.21	14.43
790	16.99	17.95	16.88	14.54

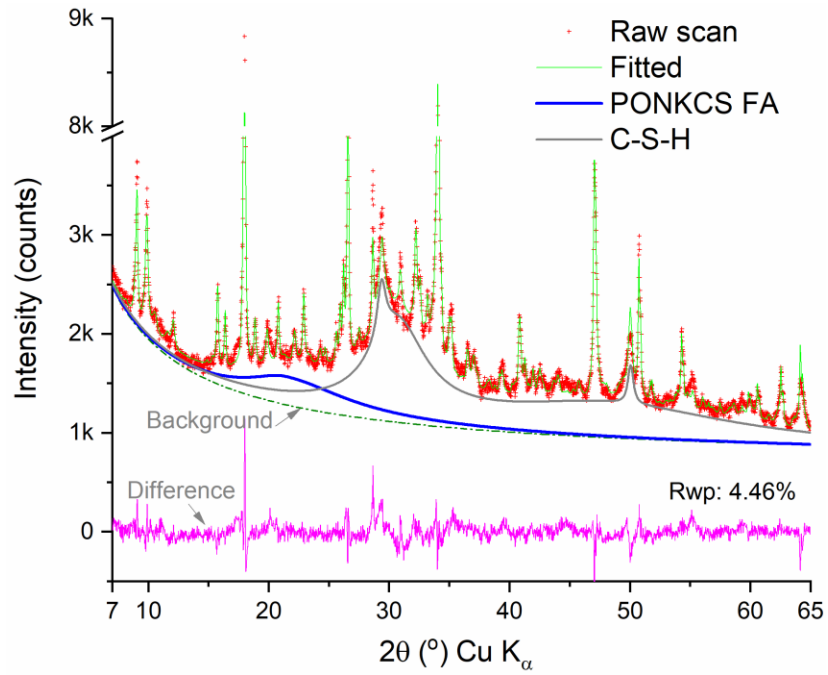


Figure S1 The fitting of the FA\_30 at 90 days, Rietveld analysis was carried out starting from 7 degrees.

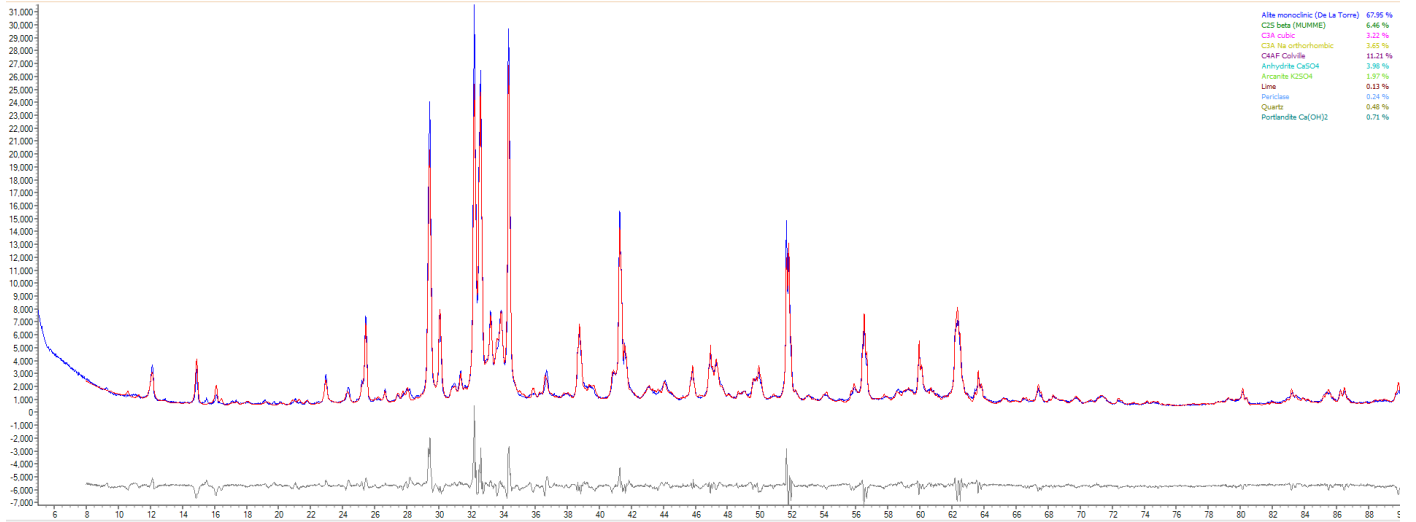


Figure S2 The Rietveld plot of the cement with a wide 2 theta angle.

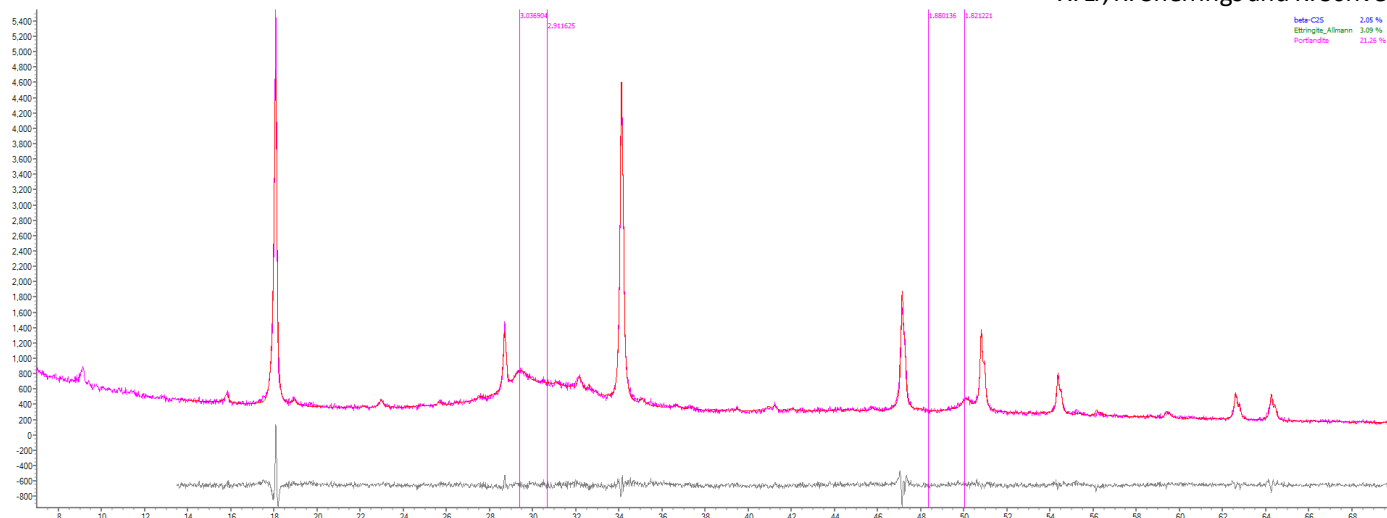


Figure S3 7-year white cement, C-S-H profile modelled using 4 peaks.

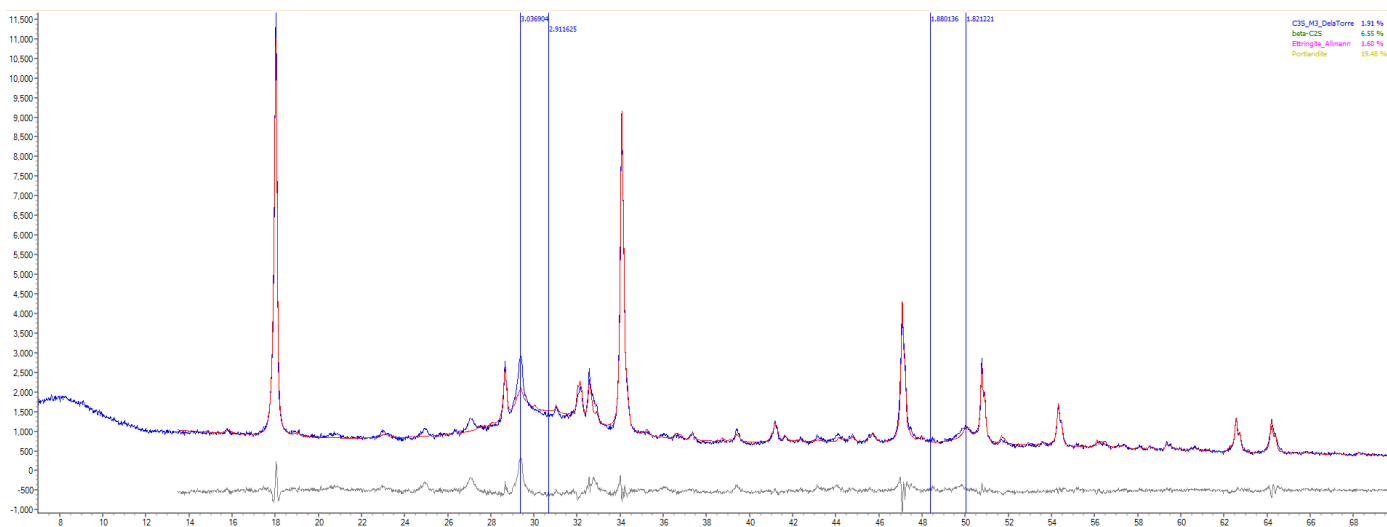


Figure S4 90d white cement, C-S-H profile from 7-year old sample

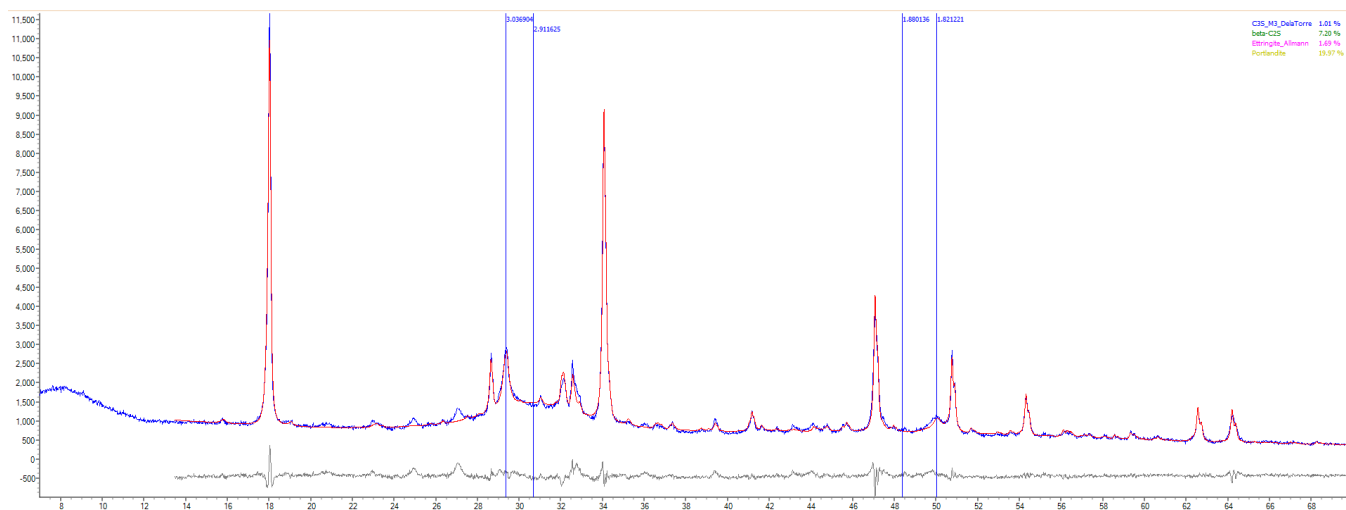


Figure S5 90d white cement, crystal size of peak at 0.3037 nm was refined